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US DEPARTMENT OF COMMERCE
PATENT AND TRADEMARK OFFICE

ATTORNEYS DOCKET NUMBER
P99,0708

**TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371**

U.S. APPLICATION NO. (if known, see 37 CFR 1.5)

09/284222

INTERNATIONAL APPLICATION NO.

INTERNATIONAL FILING DATE

PRIORITY DATE CLAIMED

PCT/JP98/03578

August 11, 1998

August 11, 1997

TITLE OF INVENTION

"NONAQUEOUS-ELECTROLYTE SECONDARY BATTERY"

APPLICANT(S) FOR DO/EO/US

Tsujimoto et al.

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay.
4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ☒ A copy of International Application as filed (35 U.S.C. 371(c)(2))
 - a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☐ has been transmitted by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US)
6. ☒ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
 - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☐ have been transmitted by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☒ have not been made and will not be made.
8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). **(UNEXECUTED)**
10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11. to 16. below concern other document(s) or information included:

11. ☐ An Information Disclosure Statement under 37 C.F.R. 1.97 and 1.98; **(PTO 1449, Prior Art, Search Report)**.
12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 C.F.R. 3.28 and 3.31 is included. (See attached envelope)
13. ☒ A **FIRST** preliminary amendment.
 - ☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
14. ☐ A substitute specification.
15. ☐ A change of power of attorney and/or address letter.
16. ☒ Other items or information:
 - a. ☒ **EXPRESS MAIL NO.:** EL270381669US dated April 9, 1999.

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17. ■ The following fees are submitted:

BASIC NATIONAL FEE (37 C.F.R. 1.492(a)(1)-(5):

Search Report has been prepared by the EPO or JPO \$840.00

International preliminary examination fee paid to USPTO (37 C.F.R. 1.482) .. \$670.00

No international preliminary examination fee paid to USPTO (37 C.F.R. 1.482) but
international search fee paid to USPTO (37 C.F.R. 1.445(a)(2)) \$760.00Neither international preliminary examination fee (37 C.F.R. 1.482) nor international
search fee (37 C.F.R. 1.445(a)(2)) paid to USPTO \$970.00International preliminary examination fee paid to USPTO (37 C.F.R. 1.482) and all
claims satisfied provisions of PCT Article 33(2)-(4) \$ 96.00**ENTER APPROPRIATE BASIC FEE AMOUNT =**

CALCULATIONS

PTO USE ONLY

\$840.00

Surcharge of \$130.00 for furnishing the oath or declaration later than ☐ 20 ☐ 30 months
from the earliest claimed priority date (37 C.F.R. 1.492(e)).

\$

Claims	Number Filed	Number Extra	Rate
Total Claims	6 - 20 =	0	X \$ 18.00
Independent Claims	1 - 3 =	0	X \$ 78.00
Multiple Dependent Claims			\$260.00 +
TOTAL OF ABOVE CALCULATIONS =			\$0.00
Reduction by 1/2 for filing by small entity, if applicable. Verified Small Entity statement must also be filed. (Note 37 C.F.R. 1.9, 1.27, 1.28)			\$
SUBTOTAL =			\$
Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).			\$
TOTAL NATIONAL FEE =			\$840.00
Fee for recording the enclosed assignment (37 C.F.R. 1.21(h). The assignment must be accompanied by an appropriate cover sheet (37 C.F.R. 3.28, 3.31). \$40.00 per property			\$
TOTAL FEES ENCLOSED =			\$840.00
			Amount to be refunded \$
			charged \$

\$0.00

\$0.00

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\$840.00

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\$840.00

a. ■ A check in the amount of \$ 840.00 to cover the above fees is enclosed.b. ☐ Please charge my Deposit Account No. _____ in the amount of \$ _____ to cover the above fees.
A duplicate copy of this sheet is enclosed.c. ■ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any
overpayment to Deposit Account No. 08-2290. A duplicate copy of this sheet is enclosed.NOTE: Where an appropriate time limit under 37 C.F.R. 1.494 or 1.495 has not been met, a petition to revive (37 C.F.R. 1.137(a) or (b)) must be
filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

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SIGNATURE

Michael R. Hull
NAME35,902
Registration Number

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re International Application of

International Serial No. PCT/JP98/03578
International filing date: August 11, 1998
For: NONAQUEOUS-ELECTROLYTE SECONDARY BATTERY

VERIFICATION OF TRANSLATION

Honorable Commissioner of Patents and Trademarks
Washington, D.C. 20231

Sir:

Eiichi Tamura, a member of A.KOIKE & CO., of 11-Mori
Bldg., 6-4, Toranomom 2-chome, Minato-ku, Tokyo 105-0001, Japan,
declares:

(1) that he knows well both the Japanese and English
languages;

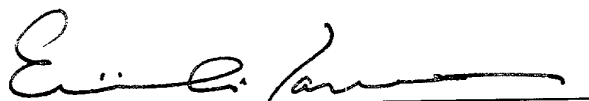
(2) that he translated the above-identified International
Application from Japanese to English;

(3) that the attached English translation is a true and
correct translation of the above-identified International
application to the best of his knowledge and belief; and

(4) that all statements made of his own knowledge are
true and that all statements made on information and belief are
believed to be true, and further that these statements are made
with the knowledge that willful false statements and the like are
punishable by fine or imprisonment, or both, under 18USC 1001,
and that such false statements may jeopardize the validity of the
application or any patent issuing thereon.

March 30, 1999

Date



Eiichi Tamura

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BOX PCT

IN THE UNITED STATES ELECTED OFFICE
OF THE UNITED STATES PATENT AND TRADEMARK OFFICE
UNDER THE PATENT COOPERATION TREATY-CHAPTER II

AMENDMENT "A" PRIOR TO ACTION

APPLICANT: Tsujimoto et al.
ATTORNEY DOCKET NO.: P99,0708
INTERNATIONAL APPLICATION NO.: PCT/JP98/03578
INTERNATIONAL FILING DATE: August 11, 1998
INVENTION: "NONAQUEOUS-ELECTROLYTE SECONDARY BATTERY"

Assistant Commissioner for Patents
Washington, D.C. 20231

SIR:

Please amend the above-identified patent application as follows, prior to examination on the merits.

IN THE SPECIFICATION

At page 1, line 1, cancel "DESCRIPTION" and substitute --TITLE-- therefor;

At page 1, line 2, cancel "NONAQUEOUS-ELECTROLYTE SECONDARY BATTERY" and substitute --"NONAQUEOUS-ELECTROLYTE SECONDARY BATTERY"-- therefor;

At page 1, line 3, cancel "TECHNICAL FIELD" and substitute --FIELD OF THE INVENTION-- therefor;

At page 1, line 7, cancel "BACKGROUND ART" and substitute --BACKGROUND OF THE INVENTION-- therefor;

At page 3, line 9, cancel "DISCLOSURE" and substitute --SUMMARY-- therefor;

At page 4, after line 11, insert the following text:

--Other objects and advantages of the present invention will become apparent from reading the following detailed description and appended claims, and upon reference to the accompanying drawings.--;

At page 4, after line 14, insert the following text:

--It should be understood that the drawings are not necessarily to scale and that the embodiments are sometimes illustrated by graphic symbols, phantom lines, diagrammatic representations and fragmentary views. In certain instances, details which are not necessary for an understanding of the present invention or which render other details difficult to perceive may have been omitted. It should be understood, of course, that the invention is not necessarily limited to the particular embodiments illustrated herein.--;

At page 4, line 15, cancel "BEST MODE FOR CARRYING OUT THE INVENTION" and substitute --**DETAILED DESCRIPTION OF THE PRESENTLY PREFERRED EMBODIMENTS**-- therefor;

At page 23, line 4, between "bodying" and "agent", insert --or reducing--;

At page 28, after line 12, insert the following text:

--From the above description, it is apparent that the objects of the present invention have been achieved. While only certain embodiments have been set forth, alternative embodiments and various modifications will be apparent from the above description to those skilled in the art. These and other alternatives are considered equivalents and within the spirit and scope of the present invention.--.

IN THE CLAIMS

Please cancel Claims 1-6 as filed in favor of new Claims 7-12 as follows:

7. A nonaqueous electrolyte secondary battery comprising:

a negative electrode comprising a binder, the binder comprising a mixture of a fluorine polymer and an aeromatic vinyl-conjugate diene polymer.

8. The nonaqueous electrolyte secondary battery of Claim 7 wherein the fluorine polymer comprises at least one material selected from the group consisting of polyvinylidene fluoride, polytetrafluoroethylene and fluorine rubber.

9. The nonaqueous electrolyte secondary battery of Claim 7 wherein the aeromatic vinyl-conjugate diene polymer comprises styrene-butadiene latex.

10. The nonaqueous electrolyte secondary battery of Claim 7 wherein the binder comprises a weight mixture ratio of the fluorine polymer to the aeromatic vinyl-conjugate diene polymer that ranges from about 1 to about 99.

11. The nonaqueous electrolyte secondary battery of Claim 7 wherein the binder comprises from about 2 weight percent to about 15 weight percent of a total weight of the negative electrode.

12. The nonaqueous electrolyte secondary battery of Claim 7 wherein the binder comprises a cellulose derivative as a viscosity reducing agent.

REMARKS

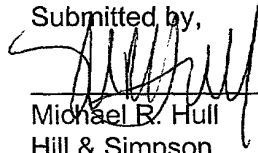
The above amendments are solely editorial and are made only for the purpose of conforming the present PCT application to the requirements of United

States patent practice. No new matter is added thereby.

The cancellation of Claims 1-6 in favor of new Claims 7-12 is done solely because amending Claims 1-6 by brackets and underlining would be extremely cumbersome in view of the large number of editorial changes which are necessary. The cancellation of Claims 1-6, therefore, is not intended to constitute a surrender of any of the subject matter of those claims, all of which is still being claimed in Claims 7-12.

An early action on the merits is respectfully requested.

Submitted by,



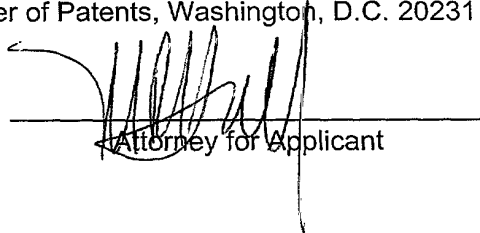
(Reg. No. 35,902)

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Attorneys for the Applicant

CERTIFICATE OF MAILING

I hereby certify that this correspondence is being deposited with the United States Postal Service as Express Mail No. EL270381669US in an envelope addressed to Assistant Commissioner of Patents, Washington, D.C. 20231 on April 8, 1999.

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Attorney for Applicant

DESCRIPTION

NONAQUEOUS-ELECTROLYTE SECONDARY BATTERY

TECHNICAL FIELD

The present invention relates to a nonaqueous electrolyte secondary battery having a large capacity and a low self-discharge ratio.

BACKGROUND ART

In recent years, wide use of portable apparatuses, such as video cameras, raises a need for secondary batteries to be employed in place of primary batteries which are disposable batteries because the secondary batteries can repetitively be used. A major portion of secondary batteries available at present are nickel-cadmium batteries each containing an alkali electrolyte. The foregoing batteries each having a voltage of about 1.2 V, however, encounter a difficulty in raising the energy density. Moreover, there arises a problem of an excessively high self-discharge ratio of 20 % or higher per month.

Therefore, a nonaqueous electrolyte secondary battery has been investigated which contain a nonaqueous solvent to serve as the electrolyte and a light metal, such as lithium, to serve as the negative electrode. Since a high voltage of 3 V or higher can be realized, the foregoing battery has a high energy density and a low self-discharge ratio. However, the above-mentioned secondary battery encounters undesirable contact of metal lithium or the like employed to form the negative electrode with the positive electrode because the metal lithium or the like is grown to a dendrite form owing to repetitive

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charging and discharging. As a result, short circuit takes place in the inside portion of the battery and the lifetime of the battery is too short to be put to practical use.

Therefore, a nonaqueous electrolyte secondary battery has been investigated in which lithium or the like is alloyed with another metal to use the obtained alloy as the negative electrode of the battery. Also in the foregoing case, the alloy is formed into very fine particles and, therefore, a problem arises in that the lifetime is shortened.

To overcome the above-mentioned problems, a nonaqueous electrolyte secondary battery has been disclosed in, for example, Japanese Patent Laid-Open No. 62-90863. In the foregoing battery, a carbonaceous material, such as cokes, is employed as an active material for the negative electrode. Since the foregoing secondary battery is free from the above-mentioned problems of the negative electrode, an excellent lifetime characteristic against cycle operation can be realized. As suggested by the inventors of the present invention in Japanese Patent Laid-Open No. 63-135099, use of Li_xMO_2 (where M is one or more types of transition metals and x is not less than 0.05 nor more than 1.10) enables a nonaqueous electrolyte secondary battery having an elongated lifetime and a high energy density to be obtained.

However, also the above-mentioned nonaqueous electrolyte secondary battery containing carbon material to serves as the active material for the negative electrode having the high energy density encounters rapid rise in the temperature if an external short circuit

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takes place in a charged state. Therefore, adverse influences are sometimes exerted on the adjacent electric elements.

The nonaqueous electrolyte secondary battery, which contains the carbonaceous material to serve as the active material for the negative electrode, exhibits a long cycle lifetime and excellent safety as compared with batteries of a type containing metal lithium or the like. However, an improvement in the cycle characteristic even under a heavy load has been required.

DISCLOSURE OF THE INVENTION

An object of the present invention is to provide a nonaqueous electrolyte secondary battery which is capable of preventing excessive rise in the temperature even if an unintentional external short circuit occurs. Another object of the present invention is to provide a nonaqueous electrolyte secondary battery having an excellent cycle characteristic even under a heavy load.

As a result of energetic investigations performed by the inventors of the present invention, a fact has been found that rise in the temperature occurring when external short circuit takes place can be prevented when a mix of fluorine polymers and aromatic vinyl-conjugate diene polymers is employed in an electrode incorporating a carbonaceous material, a collector and a binder.

That is, a nonaqueous electrolyte secondary battery according to the present invention comprises a binder for powder in a negative electrode, wherein the binder for powder in the negative electrode is a mix of fluorine polymer and aromatic vinyl-conjugate diene polymer.

The nonaqueous electrolyte secondary battery according to the present invention comprises the binder which is the mix of the fluorine polymer and the aromatic vinyl-conjugate diene polymer. Therefore, a capacity required as the battery can be maintained. Moreover, rise in the temperature occurring when an external short circuit takes place can be prevented.

Moreover, the binder for powder contains cellulose derivative to serve as a viscosity bodying agent so that prevention of rise in the temperature occurring when external short circuit takes place and improvement in the cycle characteristic even under a heavy load are realized.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 a side cross sectional view showing a nonaqueous electrolyte secondary battery.

BEST MODE FOR CARRYING OUT THE INVENTION

A nonaqueous electrolyte secondary battery according to the present invention incorporates a negative electrode, a positive electrode and a nonaqueous electrolyte.

The negative electrode is composed of an active material for the negative electrode, a binder and a collector.

A carbonaceous material for preparing the active material for the negative electrode is not limited particularly. A material, such as petroleum pitch, polymer resin or green cokes, containing resin components is a suitable material. Moreover, any one of the following materials may be employed: graphite which has completely been

carbonized, pyrolytic carbon, cokes (coal cokes, pitch cokes or petroleum cokes), carbon black (acetylene black or the like), vitreous carbon, a calcinated organic polymer material (which is obtained by calcinating an organic polymer material at appropriate temperatures not lower than 500°C in an inactive gas flow or in a vacuum), pitch containing carbon fiber and the foregoing resin and resin which can easily be sintered, for example, furan resin, divinyl benzene, polyvinylidene fluoride or vinylidene chloride and with which a mix is prepared, followed by calcinating the mix.

The present invention is characterized in that the binder in the negative electrode is a mix of fluorine polymers and aromatic vinyl-conjugate diene polymer.

When the mix of the fluorine polymer and the aromatic vinyl-conjugate diene polymer is employed as the binder, a capacity required for the battery can be maintained. Moreover, rise in the temperatures of the battery occurring when external short circuit takes place can be prevented. The excellent adhesiveness of the aromatic vinyl-conjugate diene polymer contributes to prevention of the rise in the temperature. When the aromatic vinyl-conjugate diene polymer is mixed with the fluorine polymer, a required capacity is maintained.

For example, one or more types of the following fluorine polymers may be employed: polytetrafluoroethylene, fluorine rubber, tetrafluoroethylene propylene rubber, silicon fluoride rubber, ethylene tetrafluoroperfluoroalkylvinyl ether resin (PFA), ethylene

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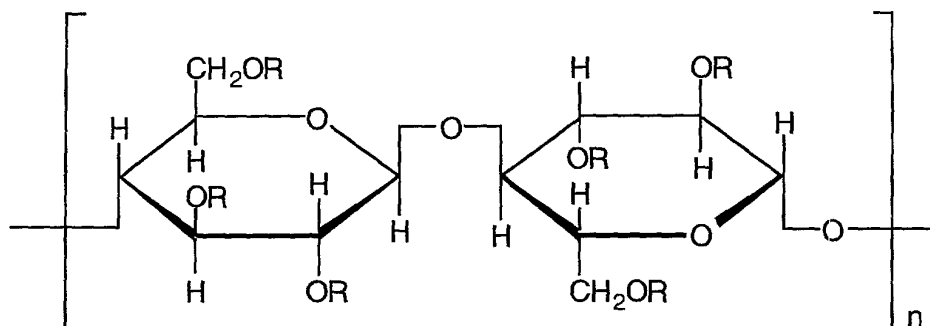
tetrafluoroethylene copolymer resin (ETFE), polychlorotrifluoroethylene (PCTFE) and tetrafluoroethylene hexafluoropropylene copolymer (FEP). The aromatic vinyl-conjugate diene polymer may be styrene-butadiene latex or the like. In addition to usual materials, a material having a hydrogenated terminal, a material having an end modified with a carboxyl group or the like may be employed to obtain a similar effect.

It is preferable that the weight mixture ratio of the fluorine polymer and the aromatic vinyl-conjugate diene polymer is set such that fluorine polymer is not less than one time nor more than 99 times the aromatic vinyl-conjugate diene polymer. The fluorine polymer cannot solely prevent rise in the temperature occurring when short circuit takes place. On the other hand, use of only the aromatic vinyl-conjugate diene polymer results in reduction in the capacity of the battery.

It is preferable that the ratio of the binder for powder with respect to the negative electrode is not less than 2 wt% nor more than 15 wt%. If the content of the binder does not satisfy the foregoing range, the capacity of the battery is undesirably reduced.

It is preferable that the binder for powder contains a cellulose derivative which serves as a viscosity bodying agent.

The cellulose derivative is expressed by the following chemical formula and classified into various types depending on the substitutional group R.



where R is one or more groups selected from H, CH_2COONa , CH_2COOLi , $\text{CH}_2\text{CH}_2\text{OH}$ [$\text{CH}_2\text{CH}(\text{CH}_3)_2\text{O}$] $_m\text{H}$ (m is an integer not smaller than 1) and CH_2COOH and n is 10 to 10,000.

In this embodiment, the cellulose derivative may be any one of the following materials: methyl cellulose, ethyl cellulose, carboxymethylcellulose (R in the foregoing formula is CH_2COONa or CH_2COOLi), carboxymethylethyl cellulose and hydroxyethyl cellulose (R in the foregoing formula is $\text{CH}_2\text{CH}_2\text{OH}$) and hydroxypropyl cellulose (R in the foregoing formula is [$\text{CH}_2\text{CH}(\text{CH}_3)_2\text{O}$] $_m\text{H}$).

When the cellulose derivative serving as the viscosity bodying agent is contained in the binder, rise in the temperature of the battery occurring when external short circuit takes place can be prevented. Moreover, the cycle characteristic even under a heavy load can be improved. It can be considered that the reason for this lies in a fact that a high liquid retention characteristic of the cellulose derivative contributes to the foregoing effect.

It is preferable that the ratio of the cellulose derivative with respect to the total weight of the negative electrode is not less than 0.1 wt% nor more than 5 wt%. If the content of the cellulose

derivative is lower than 0.1 wt%, a satisfactory effect of improving the cycle characteristic under a heavy load cannot be obtained. If the content of the cellulose derivative is higher than 5 wt%, the quantity of the active material is undesirably and correspondingly reduced.

On the other hand, the positive electrode has an active material which contains lithium composite oxide Li_xMO_2 (where M is one or more types of transition metals, preferably any one of Co, Ni and Fe and x satisfies a range $0.005 \leq x \leq 1.10$). The active material is exemplified by a lithium composite oxide expressed by LiCoO_2 , LiNiO_2 or $\text{LiNi}_y\text{Co}(1-y)\text{O}_2$ (where $0.05 \leq x \leq 1.10$ and $0 < y < 1$). Moreover, LiMn_2O_4 may be employed.

The lithium composite oxide can be obtained by mixing a carbonate, such as lithium, cobalt or nickel, in accordance with the composition, and then the mix is calcinated at temperatures in a range from 600°C to 1000°C . Note that the starting material is not limited to the carbonate. The synthesis from hydride or oxide is permitted.

The nonaqueous electrolyte may be an arbitrary material, such as a nonaqueous electrolyte obtained by dissolving an electrolyte in solvent, a material obtained by gelatinizing the foregoing nonaqueous electrolyte (a gel electrolyte) or a material obtained by swelling a polymer material with the foregoing nonaqueous electrolyte (a solid electrolyte).

The nonaqueous electrolyte may be a conventional nonaqueous electrolyte of a type obtained by dissolving an electrolyte in organic solvent. Therefore, the organic solvent is exemplified by ester, such

as propylene carbonate, ethylene carbonate or γ -butyl lactone; ether such as diethylether, tetrahydrofuran, substitution tetrahydrofuran, dioxolane, pyran, its derivative, dimethoxyethane or diethoxyethane; 3-substitution-2-oxazolidinone, such as 3-methyl-2-oxazolidione; methylsulfolane; acetonitrile; and propionitrile. One of the foregoing materials may be employed or a mixture of the same may be employed.

The electrolyte may be lithium perchlorate, lithium borofluoride, lithium phosphofluoride, lithium chloroaluminate, halogenated lithium, trifluoromethane sulfonate or the like.

When the gel electrolyte or the solid electrolyte is employed, a known gelatinizing material or polymer material may be employed. For example, any one of the following materials may be employed: polyvinylidene fluoride, copolymer of vinylidene fluoride and hexafluoropropylene, polyamide, aromatic polyamide, polyolefine, polyester, polycarbonate, polyimide, poly (meta) acrylate, polyacrylonitrile and the like.

Among the foregoing polymers, the polyacrylonitrile polymer (abbreviated as PAN type polymer) having a CN group at the side chain thereof has a high dielectric constant. Therefore, a solid polymer electrolyte having a high ion conductivity can be formed. Acrylonitrile is able to copolymerize with another monomer. Thus, a copolymer with perfluoromethacrylate, various (meta) acrylate, acrylamide, acrylic acid or methacrylic acid may be employed. In particular, it is preferable that a copolymer of (meta) acrylate and a

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monomer having ethylene oxide at the side chain thereof or a propylene oxide chain is employed.

To prove the effect of the present invention, samples were manufactured and evaluated.

Sample 1

Initially, a positive electrode 2 was manufactured as follows.

A compound for the positive electrode was prepared such that 0.5 mole of lithium carbonate and 1 mole of cobalt carbonate were mixed with each other, and then the mixture was calcinated in air at 900°C for 5 hours. Thus, LiCoO_2 was obtained. Then, LiCoO_2 was pulverized so that powder having an average particle size of 10 μm was obtained. Then, LiCoO_2 was used as an active material for the positive electrode such that 91 wt% of LiCoO_2 , 6 wt% of graphite serving as a conductive material and 3 wt% of polyvinylidene fluoride serving as a binder for the positive electrode were mixed. Thus, a mix for the positive electrode was prepared, and then the mix was dispersed in N-methyl-2-pyrrolidone so as to be slurried. Obtained slurry was applied to aluminum which was a collector for the positive electrode and which had a thickness of 20 μm . Then, the slurry was dried, and then a compression molding operation using a roller pressing machine was performed. Thus, an elongated electrode was manufactured.

Then, a negative electrode 1 was manufactured as follows.

A starting material for the active material for the negative electrode was petroleum pitch. Functional groups each containing

oxygen was introduced (so-called oxygen crosslinking) by 10 % to 20 %. Then, calcination in an inactive gas was performed at 1000°C so that a non-graphitizing carbon material was obtained which had a characteristic similar to that of a vitreous carbon material. Then, 90 wt% of the obtained carbon material and 10 wt% of polyvinylidene fluoride serving as the binder for the negative electrode were mixed with each other so that a mix for the negative electrode was prepared. Then, the mix was dispersed in N-methyl-2-pyrrolidone so as to be slurried. Obtained paste was applied to two sides of copper foil having a thickness of 10 μm , and then paste was dried and compression-molded by a roller pressing machine. Thus, an elongated electrode was manufactured.

Then, as shown in FIG. 1, the thus-obtained negative electrode 1 and positive electrode 2 were sequentially laminated such that a separator 3 in the form of a fine-porous polypropylene film having a thickness of 25 μm was interposed. Then, the laminate was wound many times around a center pin 14 so that an electrode element was manufactured.

The thus-manufactured electrode element was accommodated in a battery can 5. Moreover, an insulating plate 4 was placed on each of the upper and lower electrode element. A positive-electrode lead 13 applied with an insulating tape was drawn out from a positive-pole collector 11 so as to be welded to a safety valve unit 8 conducted with a battery cover 7. Also a negative-electrode lead 12 was drawn

out from a negative-electrode collector 10 so as to be welded to the battery can 5.

Then, an electrolyte obtained by dissolving 1 mole/liter LiPF_6 in mixed solution of propylene carbonate and diethyl carbonate was injected into the battery can 5. Then, the battery can 5 was crimped through a sealing gasket 6 so that the safety valve unit 8 and the battery cover 7 were secured. As a result, a cylindrical nonaqueous electrolyte secondary battery having a diameter of 18 mm and a height of 65 mm was manufactured.

Sample 2

A similar process to that for manufacturing Sample 1 was performed except for the binder for the negative electrode which was styrene butadiene latex (hereinafter called "SBR type latex") employed in place of polyvinylidene fluoride. Thus, a cylindrical nonaqueous electrolyte secondary battery was manufactured.

Sample 3

A similar process to that for manufacturing Sample 1 was performed except for the binder for the negative electrode which was a mix of polyvinylidene fluoride and SBR type latex at a mixture ratio of 5.0:5.0. Thus, a cylindrical nonaqueous electrolyte secondary battery was manufactured.

Sample 4

A similar process to that for manufacturing Sample 1 was performed except for the binder for the negative electrode which was a mix of polyvinylidene fluoride and SBR type latex at a mixture ratio

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of 7.0:3.0. Thus, a cylindrical nonaqueous electrolyte secondary battery was manufactured.

Sample 5

A similar process to that for manufacturing Sample 1 was performed except for the binder for the negative electrode which was a mix of polyvinylidene fluoride and SBR type latex at a mixture ratio of 8.0:2.0. Thus, a cylindrical nonaqueous electrolyte secondary battery was manufactured.

Sample 6

A similar process to that for manufacturing Sample 1 was performed except for the binder for the negative electrode which was a mix of polyvinylidene fluoride and SBR type latex at a mixture ratio of 9.0:1.0. Thus, a cylindrical nonaqueous electrolyte secondary battery was manufactured.

Sample 7

A similar process to that for manufacturing Sample 1 was performed except for the binder for the negative electrode which was a mix of polyvinylidene fluoride and SBR type latex at a mixture ratio of 9.9:0.1. Thus, a cylindrical nonaqueous electrolyte secondary battery was manufactured.

Sample 8

A similar process to that for manufacturing Sample 1 was performed except for the binder for the negative electrode which was a mix of polyvinylidene fluoride and SBR type latex at a mixture ratio of 5.0:5.0 and the content of the binder for the negative electrode

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which was 1 %. Thus, a cylindrical nonaqueous electrolyte secondary battery was manufactured.

Sample 9

A similar process to that for manufacturing Sample 1 was performed except for the binder for the negative electrode which was a mix of polyvinylidene fluoride and SBR type latex at a mixture ratio of 5.0:5.0 and the content of the binder for the negative electrode which was 2 %. Thus, a cylindrical nonaqueous electrolyte secondary battery was manufactured.

Sample 10

A similar process to that for manufacturing Sample 1 was performed except for the binder for the negative electrode which was a mix of polyvinylidene fluoride and SBR type latex at a mixture ratio of 5.0:5.0 and the content of the binder for the negative electrode which was 5 %. Thus, a cylindrical nonaqueous electrolyte secondary battery was manufactured.

Sample 11

A similar process to that for manufacturing Sample 1 was performed except for the binder for the negative electrode which was a mix of polyvinylidene fluoride and SBR type latex at a mixture ratio of 5.0:5.0 and the content of the binder for the negative electrode which was 15 %. Thus, a cylindrical nonaqueous electrolyte secondary battery was manufactured.

Sample 12

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A similar process to that for manufacturing Sample 1 was performed except for the binder for the negative electrode which was a mix of fluorine rubber (vinylidene fluoride-propylene hexafluoride copolymer) and SBR type latex at a mixture ratio of 5.0:5.0. Thus, a cylindrical nonaqueous electrolyte secondary battery was manufactured.

Sample 13

A similar process to that for manufacturing Sample 1 was performed except for carboxymethyl cellulose (expressed as "CMC" in the table) which was added as the viscosity bodying agent for the negative electrode by 0.1 wt%. Thus, a cylindrical nonaqueous electrolyte secondary battery was manufactured.

Sample 14

A similar process to that for manufacturing Sample 1 was performed except for carboxymethyl cellulose which was added as the viscosity bodying agent for the negative electrode by 1.0 wt%. Thus, a cylindrical nonaqueous electrolyte secondary battery was manufactured.

Sample 15

A similar process to that for manufacturing Sample 1 was performed except for carboxymethyl cellulose which was added as the viscosity bodying agent for the negative electrode by 3.0 wt%. Thus, a cylindrical nonaqueous electrolyte secondary battery was manufactured.

Sample 16

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A similar process to that for manufacturing Sample 1 was performed except for carboxymethyl cellulose which was added as the viscosity bodying agent for the negative electrode by 5.0 wt%. Thus, a cylindrical nonaqueous electrolyte secondary battery was manufactured.

Sample 17

A similar process to that for manufacturing Sample 1 was performed except for carboxymethyl cellulose which was added as the viscosity bodying agent for the negative electrode by 10 wt%. Thus, a cylindrical nonaqueous electrolyte secondary battery was manufactured.

Sample 18

A similar process to that for manufacturing Sample 1 was performed except for hydroxyethyl cellulose (expressed as "HEC" in the table) which was added as the viscosity bodying agent for the negative electrode by 1.0 wt%. Thus, a cylindrical nonaqueous electrolyte secondary battery was manufactured.

Characteristic Test

The thus-manufactured cylindrical nonaqueous electrolyte secondary battery according to Samples 1 to 12 were charged and discharged under the following conditions. Thus, the initial capacity and rise in the temperature occurring when external short circuit takes place in a charged state were measured. Results were shown in Tables 1 to 3.

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The thus-manufactured cylindrical nonaqueous electrolyte secondary battery according to Samples 1 and Samples 13 to 18 were charged and discharged 100 cycles under the following conditions. Thus, the initial capacity and a capacity retention ratio at the 100 cycles were measured. The discharging condition at this time corresponded to a 0.5-hour discharge ratio. That is, the foregoing test was a cycle test of discharge under a heavy load. Results were shown in Tables 4 and 5.

Conditions:

Charging: 4.2 V max. 1 A constant current

(charged with a constant current until 4.2 V and charged with a constant voltage after the voltage was raised to 4.2 V).

Discharging: 3.0 V cutoff, 0.7 A constant current

External resistance: 20 m Ω

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[Table 1]

	Content of Binder (wt%)	Mixture Ratio of Binder (with respect to 10 which was the overall quantity)		Initial Capacity (mAh)	Highest Temperature At Short Circuit (°C)
		Ratio of PVDF	Ratio of SBR		
Sample 3	10	5.0	5.0	1349	69
Sample 4	10	7.0	3.0	1350	71
Sample 5	10	8.0	2.0	1352	70
Sample 6	10	9.0	1.0	1351	76
Sample 7	10	9.9	0.1	1351	83
Sample 1	10	10.0	0.0	1353	95
Sample 2	10	0.0	10.0	1206	66

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[Table 2]

	Content of Binder (wt%)	Mixture Ratio of Binder (with respect to 10 which was the overall quantity)		Initial Capacity (mA h)	Highest Temperature At Short Circuit (°C)
		Ratio of PVDF	Ratio of SBR		
Sample 9	2	5.0	5.0	1244	58
Sample 10	5	5.0	5.0	1327	68
Sample 3	10	5.0	5.0	1349	69
Sample 11	15	5.0	5.0	1311	67
Sample 8	1	5.0	5.0	673	40

[Table 3]

	Content of Binder (wt%)	Mixture Ratio of Binder (with respect to 10 which was the overall quantity)		Initial Capacity (mA h)	Highest Temperature At Short Circuit (°C)
		Ratio of Fluorine Rubber	Ratio of SBR		
Sample 12	10	5.0	5.0	1296	65

[Table 4]

	Content of CMC (wt%)	Content of Binder (wt%)	Mixture Ratio of Binder (with respect to 10 which was the overall quantity)		Initial Capacity (mAh)	Capacity Retention Ratio (%)
			Ratio of PVDF	Ratio of SBR		
Sample 3	0	10	5	5	1349	83
Sample 13	0.1	10	5	5	1350	85
Sample 14	1	10	5	5	1339	88
Sample 15	3	10	5	5	1321	87
Sample 16	5	10	5	5	1303	88
Sample 14	10	10	5	5	1227	87

[Table 5]

	Content of HEC (wt%)	Content of Binder (wt%)	Mixture Ratio of Binder (with respect to 10 which was the overall quantity)		Initial Capacity (mAh)	Capacity Retention Ratio (%)
			Ratio of PVDF	Ratio of SBR		
Sample 18	1	10	5	5	1336	88

As can be understood from Table 1, batteries (Samples 3 to 7) in which the mixture ratio of polyvinylidene fluoride and SBR type latex

constituting the binder was 5.5 to 9.9:0.1 when the content of the binder with respect to the negative electrode was 10 wt% had large initial capacities of 1349 mAh to 1352 mAh. Moreover, a low highest temperature of 69°C to 83°C at the short circuit was observed. Thus, the foregoing batteries had structures suitable for the binder. The battery (Sample 1) containing the binder composed of only polyvinylidene fluoride had a large initial capacity of 1353 mAh. However, the highest temperature at the short circuit was 95°C which was an excessively high temperature. The battery (Sample 2) containing the binder composed of only SBR type latex had a low highest temperature at the short circuit of 66°C. However, the initial capacity was too small which was 1206 mAh.

As can be understood from Table 2, the batteries (Sample 3 and Samples 9 to 11) in which the binder was contained by 2 wt% to 15 wt% with respect to the negative electrode when the mixture ratio of polyvinylidene fluoride and SBR type latex was 5.0:5.0 had large initial capacities of 1244 mAh to 1349 mAh. Moreover, low highest temperatures of 58°C to 69°C at short circuit were realized as the structure of the binder. On the other hand, the battery (Sample 8) containing the binder by 1 wt% had a low highest temperature of 40°C at short circuit. However, the initial capacity was 673 mAh was too small.

As can be understood from Table 3, also the battery (Sample 12) containing the binder composed of fluorine rubber and SBR type latex (at a mixture ratio: 5.0:5.0) had a large initial capacity of 1296

mAh. Moreover, a low highest temperature at short circuit of 65°C was realized. Therefore, the foregoing structure was suitable for the binder.

As can be understood from Table 4, the battery (Samples 13 to 16) containing the viscosity bodying agent was carboxymethyl cellulose added by 0.1 wt% to 5 wt% had large initial capacities of 1303 mAh to 1350 mAh. As compared with the battery (Sample 3) to which carboxymethyl cellulose was not added, large capacity retention ratios of 85 % to 88 % were realized. Thus, the cycle characteristic under a heavy load was improved. On the other hand, the battery (Sample 17) containing the viscosity bodying agent which was carboxymethyl cellulose by 10 wt% encountered reduction in the active material correspondingly. Therefore, a too small initial capacity of 1227 mAh was realized.

As can be understood from Table 5, the battery (Sample 18) containing the viscosity bodying agent which was hydroxyethyl cellulose added by 1.0 wt% had a large initial capacity of 1336 mAh and a high capacity retention ratio of 88 %. Therefore, the cycle characteristic under a heavy load was improved.

As can be understood from the above-mentioned results, the structure according to the present invention is able to prevent rise in the temperature occurring when short circuit takes place while a satisfactorily large capacity is being maintained. The reason for this can be considered that the excellent adhesiveness of SBR type latex contributes to prevention of the rise in the temperature. Moreover,

mixture with polyvinylidene fluoride enables a large capacity to be maintained.

Since cellulose derivative, such as carboxymethyl cellulose or hydroxyethyl cellulose, serving as the viscosity bodying agent is added to the binder, the cycle characteristic under a heavy load can be improved. The reason for this is considered that the satisfactory liquid retention characteristic of the cellulose derivative contributes to the improvement.

Sample 19

In this example, graphite was employed in the negative electrode.

That is, the negative electrode of Sample 1 was changed to artificial graphite so that Sample 19 was manufactured.

The active material for the negative electrode was artificial graphite obtained by processing pitch cokes at a temperature of 2800°C. The obtained artificial graphite was in the form of particles having an average particle size of 30 μm , Lc of 85 and d002 of 3.336 Å.

Then, 90 wt% of the obtained carbon material, 5 wt% of polyvinylidene fluoride serving as the binder and 5 wt% of SBR type latex were mixed. Then, 1 wt% of CMC (carboxymethyl cellulose) was added so that a mix for the negative electrode was prepared. Then, the mix was dispersed in N-methyl-2-pyrrolidone so as to be slurried.

Obtained slurry was applied to the two sides of rolled copper foil serving as a collector for the negative electrode and having a thickness of 10 μm . Then, the slurry was dried, and then a compression molding operation using a roller pressing machine was performed. Thus, an elongated electrode was manufactured.

The thus-manufactured elongated electrode was employed. Moreover, a mix was obtained by dissolving, at a ratio of 1.5 moles/liter, LiPF_6 in solvent obtained by mixing ethylene carbonate, propylene carbonate, diethyl carbonate and dimethyl carbonate at ratios 2:1:2:5. The mix was employed as the electrolyte. The other operations similar to those for manufacturing Sample 1 were performed so that a battery having a diameter of 18 mm and a height of 65 mm was manufactured.

Sample 20

In this example, the viscosity bodying agent was hydroxypropyl cellulose.

A process similar to that for manufacturing Sample 19 was performed to manufacture a battery except for the viscosity bodying agent which was hydroxypropyl cellulose employed in place of CMC.

Sample 21

In this example, the viscosity bodying agent was electrolytic copper foil.

A process similar to that for manufacturing Sample 19 was performed to manufacture a battery except for the collector for the negative electrode which was electrolytic copper foil which had the

same thickness as that of rolled copper foil and which was employed in place of the rolled copper foil.

Sample 22

In this example, the positive electrode was a nickel material.

Initially, the nickel type positive electrode was manufactured as follows.

A compound for the positive electrode was obtained such that 1 mole of lithium hydroxide, 0.8 mole of nickel hydroxide and 0.2 mole of cobalt hydroxide were mixed, followed by calcinating the mix at 800°C for 5 hours in an oxygen atmosphere. Thus, $\text{LiNi}_{0.8}\text{Co}_{0.2}\text{O}_2$ was obtained.

Obtained $\text{LiNi}_{0.8}\text{Co}_{0.2}\text{O}_2$ was pulverized by a bowl mill so that powder having an average particle size of 10 μm was obtained.

Then, 91 parts by weight of $\text{LiNi}_{0.8}\text{Co}_{0.2}\text{O}_2$, 6 parts by weight of graphite serving as a conductive material and 3 parts by weight of polyvinylidene fluoride serving as a binder were mixed. Then, N-methyl-pyrrolidone serving as a dispersant was added so that paste was prepared. The paste was dried, and then obtained slurry was applied to aluminum foil which was a collector for the positive electrode and which had a thickness of 20 μm . Then, the slurry was dried, and then compression molding using a roller pressing machine was performed. Thus, an elongated electrode was manufactured.

The negative electrode was manufactured similar to Sample 19 so that a battery similar to that according to Sample 19 was manufactured.

Sample 23

In this example, a material was obtained by adding another type resin component to SBR type latex.

Although the SBR according to Sample 19 had a composition that styrene : butadiene : the other resin component = 40:60:0, the composition according to this example was such that styrene : butadiene : the other resin component = 35:30:35. The other operations were similar to that for manufacturing Sample 19 so that a battery was manufactured.

Sample 24

A process similar to that for manufacturing Sample 19 was performed to manufacture a battery except for CMC which was omitted from the binder for the negative electrode of the Sample 19.

Characteristic Test

The thus-manufactured cylindrical nonaqueous electrolyte secondary batteries according to Samples 19 to 24 were charged and discharged 100 cycles under the following condition. Then, the initial capacities and capacity retention ratios after the 100 cycles were completed were measured. Results were shown in Table 6.

Conditions:

Charging: 4.2 V max. 1 A constant current

(charged with a constant current until 4.2 V and charged with a constant voltage after the voltage was raised to 4.2 V).

Discharging: 3.0 V cutoff, 3 A constant current

[Table 6]

	Cellulose Derivative Content (wt%)	Content of Binder (wt%)	Mixture Ratio of Binder (with respect to 10 which was the overall quantity)	
			Ratio of PVDF	Ratio of SBR
Sample 19	CMC 1%	10	5	5
Sample 20	HPC 1%	10	5	5
Sample 21	CMC 1%	10	5	5
Sample 22	CMC 1%	10	5	5
Sample 23	CMC 1%	10	5	5
Sample 24	0%	10	5	5

	Initial Capacity (mAh)	Cycle Retention Ratio 100 cycles/ initial state
Sample 19	1400	90
Sample 20	1400	91
Sample 21	1400	93
Sample 22	1400	92
Sample 23	1500	90
Sample 24	1350	75

As can be understood from the foregoing results, Samples 19 to 23 according to the examples of the present invention had excellent charging/discharging efficiency and cycle characteristic. The reason

why the cycle characteristic was improved as compared with Sample 24 in which CMC was not added lies in that addition of fibrous molecules, such as CMC, caused SBR and CMC to be mixed and adhesion to be performed without deterioration in the reactivity of the surfaces of the carbon portions.

Although the cylindrical nonaqueous electrolyte secondary batteries were manufactured in the foregoing examples of the present invention, similar effects can be obtained when the present invention is applied to batteries having another shape, for example, a rectangular shape, a laminate shape or a card shape.

Note that teflon or the like may be employed as the fluorine binder.

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CLAIMS:

1. A nonaqueous electrolyte secondary battery comprising: a binder for powder in a negative electrode, wherein said binder for powder contains a mix of fluorine polymer and aromatic vinyl-conjugate diene polymer.
2. A nonaqueous electrolyte secondary battery according to claim 1, wherein the fluorine polymer is at least one type of materials selected from a group consisting of polyvinylidene fluoride, polytetrafluoroethylene and fluorine rubber.
3. A nonaqueous electrolyte secondary battery according to claim 1, wherein the aromatic vinyl-conjugate diene polymer is styrene-butadiene latex.
4. A nonaqueous electrolyte secondary battery according to claim 1, wherein a weight mixture ratio of fluorine polymer and aromatic vinyl-conjugate diene polymer is such that fluorine polymer is not less than one time nor more than 99 times the aromatic vinyl-conjugate diene polymer.
5. A nonaqueous electrolyte secondary battery according to claim 1, wherein a ratio of said binder for powder with respect to the total weight of said negative electrode is not less than 2 wt% nor more than 15 wt%.
6. A nonaqueous electrolyte secondary battery according to claim 1, wherein said binder for powder contains cellulose derivative as a viscosity bodying agent.

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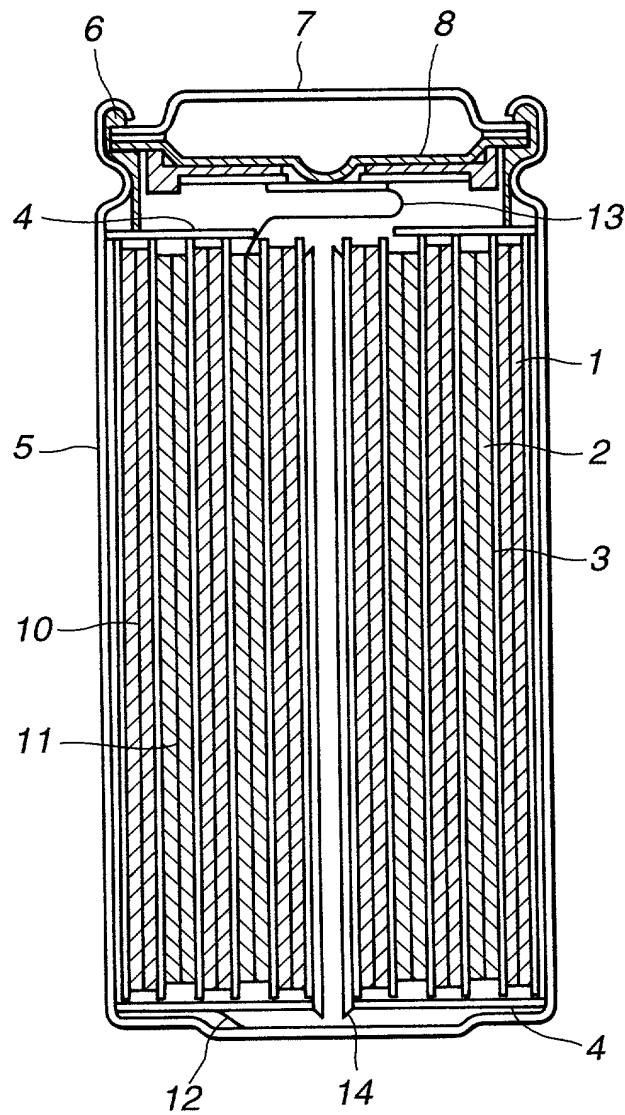


FIG.1

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DECLARATION AND POWER OF ATTORNEY

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

NONAQUEOUS-ELECTROLYTE SECONDARY BATTERY

Case No. P99,0708, the specification of which

(check one) ☒ is attached hereto.
☐ was filed on April 9, 1999, as
Application Serial No. 09/284,222
and was amended on _____
(if applicable)

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims as amended by any amendment referred to above.

I acknowledge the duty to disclose to the United States Patent Office all information which is known to me to be material to the patentability of this application in accordance with Title 37, Code of Federal Regulations, 1.56.¹

I do not know and do not believe this invention was ever known or used in the United States of America before my or our invention thereof, or patented or described in any printed publication in any country before my or our invention thereof or more than one year prior to this application, that the same was not in public use or on sale in the United States of America more than one year prior to this application, and I believe that the invention has not been patented or made the subject of an inventor's certificate issued before the date of this application in any country foreign to the United States of America on an application filed by me or my legal representatives or assigns more than twelve months prior to this application, and that no application for patent or inventor's certificate on this invention has been filed in any country foreign to the United States of America prior to this application by me or my legal representatives or assigns, except as identified below:

I hereby claim foreign priority benefits under Title 35, United States Code, 119 of any foreign application(s) for patent or inventor's certificate listed below

Prior Foreign Application(s)

Number Country

P9-216468 Japan

P9-361060 Japan

PCT/JP98/03578 PCT

Date

August 11, 1997

December 26, 1997

August 11, 1998

and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the above listed application on which priority is claimed:

Prior Foreign Application(s)

Number Country

Date

If no priority is claimed, I have identified all foreign patent applications filed prior to this application:

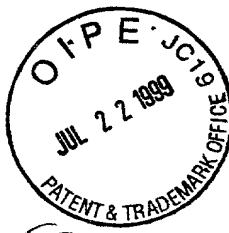
¹ (b) Under this section, information is material to patentability when it is not cumulative to information already of record or being made of record in the application, and

- (1) It establishes, by itself or in combination with other information, a *prima facie* case of unpatentability of a claim; or
- (2) It refutes, or is inconsistent with, a position the applicant takes in:

- (i) Opposing an argument of unpatentability relied on by the Office, or
- (ii) Asserting an argument of patentability.

A *prima facie* case of unpatentability is established when the information compels a conclusion that a claim is unpatentable under the preponderance of evidence, burden-of-proof standard, giving each term in the claim its broadest reasonable construction consistent with the specification, and before any consideration is given to evidence which may be submitted in an attempt to establish a contrary conclusion of patentability.

S98P10320500



21

Prior Foreign Application(s) _____
Number _____ Country _____ Date _____

And I hereby appoint Messrs. John D. Simpson (Registration No. 19,842), Dennis A. Gross (24,410), Robert M. Barrett, (30,142), Steven H. Noll (28,982), Kevin W. Gynn (29,927), Robert M. Ward (26,517), Brett A. Valiquet (27,841), Edward A. Lehman (22,312), David R. Metzger (32,919), Todd S. Parkhurst (26,494), James D. Hobart (24,149), Melvin A. Robinson (31,870), John R. Garrett (27,888), Paula J. Kelly (37,624), John W. Cornell (30,619), Robert J. Depke (37,607), Joseph P. Reagen (35,332), Michael R. Hull (35,902), Michael S. Leonard (37,557), William E. Vaughan (39,056) and Lewis T. Steadman (17,074), all members of the firm of Hill & Simpson, A Professional Corporation

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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1-00
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